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L3: Entry 1 of 2

File: USPT

Mar 16, 1993

US-PAT-NO: 5194477

DOCUMENT-IDENTIFIER: US 5194477 A

TITLE: Flame retardant compositions

DATE-ISSUED: March 16, 1993

## INVENTOR-INFORMATION:

NAME	CITY	STATE	ZIP CODE	COUNTRY
Liu; Ping Y.	Naperville	IL		
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US-CL-CURRENT: 524/382; 524/399, 524/409, 524/410, 524/412, 525/425, 525/433,  
525/66, 525/67

Full	Title	Citation	Front	Review	Classification	Date	Reference	Claims	EXMC	Draw Desc	Image
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☐ 2. Document ID: EP 838503 A1

L3: Entry 2 of 2

File: EPAB

Apr 29, 1998

PUB-NO: EP000838503A1

DOCUMENT-IDENTIFIER: EP 838503 A1

TITLE: Flame retardant composition of polyerstercarbonate, halogenated polycarbonate, potassium diphenylsulfone sulfonate and polytetrafluoroethylene

PUBN-DATE: April 29, 1998

## INVENTOR-INFORMATION:

NAME	COUNTRY
PAN, WIE-HIN	US
PATTERSON, DWIGHT JUAN	US
BOUTNI, OMAR MOHAMED	US
FONTANA, LUCA PIETRO	US

INT-CL (IPC): C08L 69/00; C08K 5/42

EUR-CL (EPC): C08L069/00; C08K005/42, C08L069/00

Full	Title	Citation	Front	Review	Classification	Date	Reference	Claims	EXMC	Draw Desc	Image
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Art Unit: 1711

applicant states, one would seek enough information from secondary references to use to modify the primary reference. The applicant on page 3, paragraph 1, contends that addition of proteins to compositions containing alcohols or others hydroxy compounds will most likely result in denaturation of proteins. This argument is not persuasive without any supportive data.

The determination whether prior art is analogous or not, involves some factual issues concerning whether a references (or preferences) is reasonably pertinent to the particular problem with which the invention was concerned. In the instant invention that is the case. Each of the four references under prior art is directed to solving the same problem as that of instant invention viz freeze stability of an emulsion based system. Prior art therefore is analogous.

Two submitted (published) articles on denaturation of proteins have been considered. Denaturation does not necessarily imply destruction. Hence applicant's argument on page 3, middle of paragraph 1 of (earlier filed) paper #5 is not persuasive.

It should be noted that a foodstuff like milk and a binder (in a point formulation) which is a latex are physically in the same form viz emulsion. Also, a protein and a polypeptide are similar to each other, both containing chains of amino acids. Therefore it is reasonable to

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L3: Entry 1 of 2

File: USPT

Mar 16, 1993

DOCUMENT-IDENTIFIER: US 5194477 A  
TITLE: Flame retardant compositions

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## ABPL:

Flame retardant polyester compositions exhibiting good processability comprising at least one polyester resin and a flame retardant additive comprising a halogenated polycarbonate resin and an antimony compound in a polyetherimide ester elastomer resin carrier.

## BSPR:

This invention relates to flame retardant thermoplastic molding compositions, particularly thermoplastic polyesters and mixtures of polyesters and polycarbonates, having incorporated therein a novel flame retardant additive for such thermoplastic compositions. More particularly, disclosed are flame retardant polyester compositions having good physical properties and exhibiting good processability comprising a polyester resin, preferably a poly(butylene terephthalate) resin; and a flame retardant amount of a novel flame retardant comprised of (1) an antimony compound; (2) a halogenated polycarbonate resin and (3) a polyetherimide ester elastomeric resin. The novel flame retardant additive of the present invention can also be utilized effectively in polyester compositions modified with a polycarbonate resin.

## BSPR:

In addition to the optional use of a polycarbonate resin in the compositions of the present invention, said compositions can also optionally advantageously contain one or more of a resinous impact enhancer and/or a mineral reinforcing agent.

## BSPR:

It has surprisingly been discovered that standard polyester flame retardant compositions, when modified with a novel flame retardant additive composition that is comprised of a halogenated polycarbonate resin, an antimony compound and a polyetherimide ester elastomer have superior processing properties than polyester compositions not modified with this novel flame retardant additive or modified with conventional flame retardant compositions. This superior processability may be shown by the lower melt viscosity exhibited by examples of the present invention over a control sample formulated with a conventional flame retardant additive. In addition, it has been discovered that various other physical properties of polyester compositions formulated with the novel concentrate of the present invention are comparable to those properties exhibited by the control samples.

## BSPR:

These polyesters may be advantageously modified by being blended with a polycarbonate resin. In addition, these resins can be blended with a resinous impact enhancer or a reinforcing agent.

## BSPR:

According to the present invention, these polyester resins are formulated with a flame retardant additive which comprises a halogenated polycarbonate resin, an antimony compound and a polyetherimide ester elastomer resin carrier.

## BSPR:

The second component of the flame retardant composition of the present invention is a halogenated polycarbonate resin.

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Date: \_\_\_\_\_

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BSPR:

Preferred halogenated polycarbonate resins are those derived from, at least in part, halogenated dihydric phenols. These include high and low molecular weight polycarbonates and copolycarbonates as well as the diglycidyl ethers of dihydric phenols. The former may be prepared by reacting the dihydric phenol with a carbonate precursor, e.g. carbonyl bromide or carbonyl chloride and, optionally, other dihydric phenols, glycols and/or dicarboxylic acids. The latter may be prepared by the reaction of dihydric phenol with a glycidyl or a glycidyl generating monomer such as epichlorohydrin. Suitable examples of the halogenated divalent phenols that may be employed in the preparation of the carbonate and the epoxy resins include 2,2-bis(3,5-dibromo-4-hydroxyphenyl) propane, 2,2-bis(3,5-dichloro-4-hydroxyphenyl) propane, bis(3,5-dibromo-4-hydroxyphenyl) methane, bis(3,5-dichloro-4-hydroxyphenyl) methane, 2,2-bis(4-hydroxy-2,3,5,6-tetrabromophenyl) propane, 2,2-bis(4-hydroxy-2,3,5,6-tetrachlorophenyl) propane, or the like. The most preferred halogenated dihydric phenol is 2,2-bis(3,5-dibromo-4-hydroxyphenyl) propane, wherein said carbonate polymer or copolymer and epoxy resin will have repeating units of formula VI and VII respectively. ##STR6## wherein Z is a halogen atom, preferably, bromine or chlorine; b and c are, independently, whole numbers, including zero, and i is a whole number from 1-4, preferably 2.

BSPR:

Preferred halogenated carbonate polymer flame retardants are (1) those having 25 to 75, preferably 30 to 45, weight percent of the repeating units comprising the chloro- or bromo-substituted dihydric phenol units and the remainder comprising dihydric phenol, glycol or dicarboxylic acid units and (2) low molecular weight tetrabromobisphenol A polycarbonates of 2 to 10, preferably 4 to 7 repeating units and end capped with tribromophenol.

BSPR:

The molecular weight of the halogenated polycarbonates may vary widely and are generally from 2000 to 40,000. A preferred high molecular weight halogenated carbonate polymer will have a molecular weight ranging from about 20,000 to about 40,000. Also, the halogen content of the halogenated carbonate material may vary widely. The preferred halogen content is from about 20% to greater than about 80% by weight, preferably from about 35% to about 70%.

BSPR:

The amount of the impact modifier can also vary widely. It has been found that even relatively minor amounts of the modifier are effective in providing significant improvements in impact strength over a range of low temperatures. The optimum amounts of optional impact modifier used will depend on the type of impact modifier employed. Generally, an impact modifier will be used in amounts ranging from about 5 to about 75 parts by weight, preferably from about 10 to about 30 parts by weight based on 100 parts by weight of the total resin component, that is, the total of polyester resin, impact modifier resin, and polycarbonate resin, if any, utilized in the composition of the present invention.

BSPR:

As indicated, the composition of the present invention may also optionally include a polycarbonate resin.

BSPR:

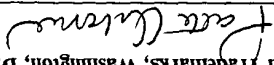
Polycarbonates suitable for use in the present invention are any of those known in the art. Especially preferred polycarbonates are high molecular weight, thermoplastic, aromatic polymers and include homopolycarbonates, copolycarbonates and copolyestercarbonates and mixtures thereof which have average molecular weights of about 8,000 to more than 200,000, preferably of about 20,000 to 80,000 and an I.V. of 0.30 to 1.0 dl/g as measured in methylene chloride at 25.degree. C. In one embodiment, the polycarbonates are derived from dihydric phenols and carbonate precursors and generally contain recurring structural units of the formula; ##STR7## where Y is a divalent aromatic radical remaining after removal of the hydroxy groups from the dihydric phenol employed in the polycarbonate producing reaction, and n is greater than 1, preferably from about 10 to about 400.

BSPR:

Preferred polycarbonate resins are of the formula: ##STR8## wherein R.sub.1 and

Honorable Commissioner of  
Patents and Trademarks  
Washington, D.C. 20231

Dear Sir:

DATE OF DEPOSIT JANUARY 26, 1995
I hereby certify that this paper or fee is being deposited with the United States Postal Service "EXPRESS MAIL, POST OFFICE TO ADDRESS" service under 37 C.F.R. 1.10 on the date indicated above and is addressed to The Commissioner of Patents and Trademarks, Washington, D.C. 20231.

Signature

This is a Request for filing a continuing application under 37 C.F.R. § 1.60 of pending prior application Serial No. 08/240,299, filed on May 9, 1994, entitled BAR CODE READER USING SCANNED MEMORY ARRAY in the names of Mark J. Krilchever and Boris Metlitsky.

The following are enclosed:

1. X Papers which the undersigned declares to be a true copy of the prior application as originally filed, including a 22 page disclosure, 10 page(s) of claims, a 1 page abstract of the disclosure, 3 sheets of informal drawings, and a declaration signed by the inventor(s).

2. X A check in the amount of \$1,550.00 to cover the filing fee for 40 total claims, 8 of which are independent which will be in the application after preliminary amendment. However, if this check is missing or is in error in amount, the Commissioner is authorized to charge any additional fees which may be required, or credit any overpayment, to Account No. 01-2508, Order No. SYMB:002-3/HAR. Two copies of this paper are enclosed for this purpose.

- 3.a. X A copy of the Power of Attorney in the prior application.

or

- 3.b.        A new Power of Attorney

4. X An assignment of record for the original application, Serial No. 07/317,533, filed March 1, 1989.

5. X Three (3) sets of informal drawings, each set consisting of 3 sheets.

- 6.a.        A verified statement claiming small entity status is enclosed.

or

- 6.b.        A verified statement claiming small entity status was filed in a parent application and small entity status is still proper and desired in this continuing application.

X Please address all correspondence in connection with this application Wayne M. Harding, Arnold, White & Durkee, P.O. Box 4433, Houston, Texas 77210; telephone (713) 787-1400.

XX Amend the specification by inserting before the first line the sentence:

--This application is a continuation under 37 C.F.R. § 1.60 of pending application Serial No. 08/240,299, filed May 9, 1994, which was a file wrapper continuation of Serial No. 08/041,281, filed March 30, 1993, which was a file wrapper continuation of Serial No. 07/317,533, filed March 1, 1989.--

R.sub.2 are independently hydrogen, (lower) alkyl or phenyl and m is at least 30, preferably between 40 and 300. The term (lower) alkyl includes hydrocarbon groups of from 1 to 6 carbon atoms.

BSPR:

Suitable dihydric phenols for producing polycarbonates include, for example, 2,2-bis(4-hydroxyphenyl)propane, bis(4-hydroxyphenyl)-methane, 2,2-bis(4-hydroxy-3-methylphenyl)propane, 4,4-bis(4-hydroxyphenyl)heptane, 2,2-(3,5,3',5'-tetrachloro-4,4'-dihydroxyphenyl)propane, 2,2(3,5,3',5'-tetrabromo-4,4'-dihydroxyphenyl)propane, and 3,3'-dichloro-4,4'-dihydroxydiphenylmethane.

BSPR:

Other dihydric phenols which are also suitable for use in the preparation of the above polycarbonates are disclosed in U.S. Pat. Nos. 2,999,835; 3,038,365; 3,169,121; 3,334,154; and 4,131,575, incorporated herein by reference.

BSPR:

It is, of course, possible to employ two or more different dihydric phenols or a copolymer of a dihydric phenol with a glycol or with a hydroxy or acid terminated polyester, or with a diacid in the event a carbonate copolymer or interpolymer rather than a homopolymer is desired for use in the preparation of the blends of the invention. Blends of any of the above materials can also be employed to provide the aromatic polycarbonate. In addition, branched polycarbonates such as are described in U.S. Pat. No. 4,001,184, can also be utilized in the practice of this invention, as can blends of a linear polycarbonate and a branched polycarbonate.

BSPR:

The polycarbonate may also be a copolyestercarbonate as described by Clayton B. Quinn in U.S. Pat. No. 4,430,484 and Kenneth Miller in U.S. Pat. No. 4,465,820 and the references cited therein, incorporated herein by reference. Preferred polyestercarbonates are those derived from the dihydric phenols and carbonate precursors described above and aromatic dicarboxylic acids or their reactive derivatives, such as the acid dihalides, e.g. dichlorides. A quite useful class of polyestercarbonates are the aromatic polyestercarbonates derived from bisphenol A; terephthalic acid or isophthalic acid or a mixture thereof or their respective acid chlorides; and phosgene. If a mixture of terephthalic acid and isophthalic acid is employed, the weight ratio of terephthalic acid to isophthalic acid may be from about 2:8 to about 8:2.

BSPR:

The polycarbonates of the subject blends can be manufactured by known processes, such as, for example, by reacting a dihydric phenol with a carbonate precursor such as diphenyl carbonate or phosgene in accordance with methods set forth in the above-cited literature and U.S. Pat. Nos. 4,018,750 and 4,123,436, or by transesterification processes such as are disclosed in the U.S. Pat. No. 3,151,008, as well as other processes known to those skilled in the art. The aromatic polycarbonates are typically prepared by employing a molecular weight regulator, an acid acceptor and a catalyst. The molecular weight regulators which can be employed include phenol, cyclohexanol, methanol, alkylated phenols, such as octylphenol, parateriary-butyl-phenol, etc. Preferably, phenol or an alkylated phenol is employed as the molecular weight regulator.

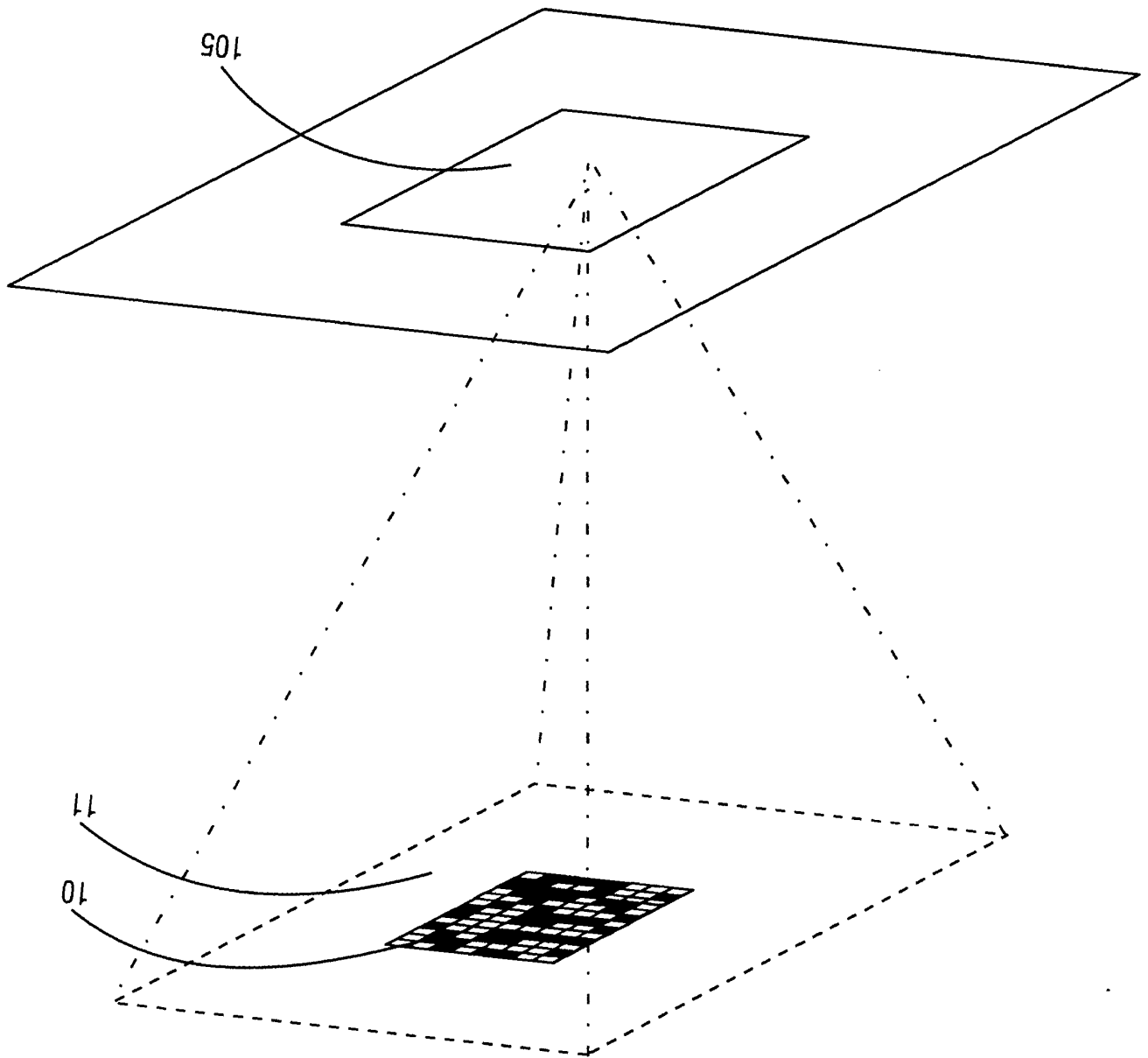
BSPR:

Also included are branched polycarbonates wherein a polyfunctional aromatic compound is reacted with the monomer and carbonate precursor to provide a thermoplastic randomly branched polycarbonate. The polyfunctional aromatic compounds contain at least three functional groups which are carboxyl, carboxylic anhydride, haloformyl, or mixtures thereof. Illustrative polyfunctional aromatic compounds which can be employed include trimellitic anhydride, trimellitic acid, trimellityl trichloride, 4-chloroformyl phthalic anhydride, promellitic acid, pyromellitic dianhydride, mellitic acid, mellitic anhydride, trimesic acid, benzophenonetetracarboxylic acid, benzophenonetetracarboxylic anhydride, and the like. The preferred polyfunctional aromatic compounds are trimellitic anhydride and trimellitic acid or their acid halide derivatives.

BSPR:

The amount of the optional polycarbonate resin utilized in the composition of the

FIG. 14





present invention can vary widely and is not critical to the present invention. Generally, the polycarbonate resin will be used in amounts ranging from about 25 to about 500 parts by weight, preferably from about 50 to about 400 parts by weight, more preferably from about 100 to about 300 parts by weight, and most preferably from about 150 to about 250 parts by weight, based on 100 parts by weight of the polyester resin component.

**BSPR:**

In the flame retardant additive, the halogenated polycarbonate resin is preferably present in amounts ranging from about 30 weight percent to about 85 weight percent, more preferably in amounts ranging from about 40 weight percent to about 75 weight, and most preferably in amounts ranging from about 60 to about 70 weight percent based on the total weight of the flame retardant additive; the antimony compound is preferably present in amounts ranging from about 5 weight percent to about 40 weight percent, more preferably from about 10 weight percent to about 35 weight percent and most preferably from about 20 weight percent to about 30 weight percent based on the total weight of the flame retardant additive; and the polyetherimide ester elastomer resin is preferably present in amounts ranging from about 10 weight percent to about 30 weight percent, more preferably in amounts ranging from about 12 weight percent to about 25 weight percent and most preferably in amount ranging from about 15 to about 20 weight percent based on the total weight of the flame retardant additive.

**DEPR:**

Examples 1 and 3 illustrate a composition falling outside the scope of the instant invention and are presented for comparative purposes only. These examples utilize the same components in their composition as the composition of, respectively, Examples 2 and 4, with the exception that the polyetherimide ester elastomer carrier of the present invention which is utilized in the flame retardant additive of Examples 2 and 4 was replaced by a different carrier, ethyl vinyl acetate carrier ("EVA") for comparative purposes. The designation FRC-1 in the Examples stands for the comparative flame retardant additive which comprises halogenated polycarbonate resin, antimony trioxide and EVA in, respectively, the following weight percentages: 67:20:13. The designation FRC-2 stands for the flame retardant additive of the present invention in which, as indicated, the same amount by weight of polyetherimide ester ("PEI") resin is substituted for EVA. The amount, by weight, of the other components in the compositions of Examples 2 and 4 also remained the same relative to, respective, the compositions of Examples 1 and 3. The PBT (poly (1,4-butylene terephthalate) utilized in the examples was Valox.RTM. 315 from General Electric Company; the halogenated polycarbonate resin utilized was BVC-58 made by Great Lakes Chemical Company, a high molecular weight brominated polycarbonate resin that has 58% brominated polycarbonate; and the impact modifier utilized in Examples 1 and 2 was Paraloid KM-653, from Rohm and Haas, and the polycarbonate utilized was General Electric Company's Lexan.RTM. polycarbonate resin. In all the Examples the resulting polymeric composition was formed, by injection molding, into test bars measuring 2.5 inches.times.1/2 inches.times.1/8 inch. A number of tests were taken of the physical properties of the composition according to the ASTM standards noted above and the test procedure set forth in Underwriters Laboratories UL94 Bulletin, and the results are set forth in Tables II and IV below. All parts in Tables I and III are listed in terms of percent by weight.

**DETL:**

TABLE I		EXAMPLE Composition 1 2			
		PBT	20.00	20.00	<u>Polycarbonate</u> 47.45 47.45
Impact Modifier	12.00 12.00	Stabilizer	1.25 1.25	Teflon Concentrate	0.30 0.30
FRC-1	19.00 --	FRC-2	--	19.00	

**CLPR:**

16. The composition of claim 1 wherein the halogenated polycarbonate compound is a brominated polycarbonate compound.

**CLPR:**

19. A composition of claim 1 which further contains a polycarbonate resin of the formula ##STR15## wherein R.sub.1 and R.sub.2 are independently hydrogen, (lower) alkyl or phenyl and m is at least 30.

**CLPR:**

20. The composition of claim 19 wherein the aromatic polycarbonate resin is

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Total (1)	1	-

derived from a diphenol and phosgene or a phosgene precursor.

CLPR:

26. A flame retardant additive which is comprised of (a) a halogenated polycarbonate resin; (b) an antimony compound; and (c) a polyetherimide ester elastomer resin carrier, wherein the halogenated polycarbonate resin is present in amounts ranging from about 60 weight percent to about 70 weight percent based on the total weight of the additive, the antimony compound is present in amounts ranging from about 20 weight percent to about 30 weight percent based on the total weight of the additive and the polyetherimide ester elastomer resin carrier is present in amounts ranging from about 15 weight percent to about 20 weight percent based on the total weight of the additive.

CLPV:

(b) a flame retardant amount of a flame retardant additive comprising a halogenated polycarbonate resin composition and an antimony compound in polyetherimide ester elastomer resin carrier, wherein, in the additive, the halogenated polycarbonate resin is present in amounts ranging from about 60 weight percent to about 70 weight percent based on the total weight of the additive, the antimony compound is present in amounts ranging from about 20 weight percent to about 30 weight percent based on the total weight of the additive and the polyetherimide ester elastomer resin carrier is present in amounts ranging from about 15 weight percent to about 20 weight percent based on the total weight of the additive.



The United States.

To all to whom these Presents shall come. Greeting.

Whereas Samuel Hopkins of the City of Philadelphia and State of Pennsylvania hath discovered an Improvement, not known or used before, such Discovery, in the making of Pot. ash and Pearl ash by a new Apparatus and Process, that is to say, in the making of Pearl ash 1<sup>st</sup> by burning the raw Ashes in a Furnace; 2<sup>d</sup> by dissolving and boiling them when so burnt in Water, 3<sup>d</sup> by drawing off and settling the ley, and 4<sup>th</sup> by boiling the ley into darts which then are the true Pearl ash, and also in the making of Pot. ash by placing the Pearl ash so made as aforesaid; which Operation of burning the raw Ashes in a Furnace, preparatory to their Dissolution and boiling in Water, is new, leaves little Residuum; and produces a much greater Quantity of Salt: These are therefore in pursuance of the Act, entitled "An Act to promote the Progress of useful Arts", to grant to the said Samuel Hopkins, his Heirs, Administrators and Assigns, for the Term of fourteen Years, the sole and exclusive Right and Liberty of using and vending to others the said Discovery of burning the raw Ashes previous to their being dissolved and boiled in Water, according to the true Intent and Meaning of the Act aforesaid. In Testimony whereof I have caused these Letters to be made patent, and the Seal of the United States to be hereunto affixed. Given under my Hand at the City of New York this thirty first Day of July in the Year of our Lord one thousand seven hundred & Ninety.

Washington

City of New York July 31<sup>st</sup> 1790. -

I do hereby certify that the foregoing Letters patent were delivered to me in pursuance of the Act, entitled "An Act to promote the Progress of useful Arts"; that I have examined the same, and find them conformable to the said Act.

Edm: Randolph Attorney General for the United States.

X000001  
July 31, 1790